

LA-UR-78-1183

MASTER

CONF-780417-9

TITLE: MASS TRANSFER CHANGES INDUCED IN COAL BLOCKS DURING THERMAL PROCESSING

AUTHOR(S): N. E. Vanderborgh, G-7
J. P. Bertino, CMB-8
D. N. Hopkins, G-7

SUBMITTED TO: 1978 Spring Meeting Western States Section/
The Combustion Institute, Boulder, CO

By acceptance of this article for publication, the publisher recognizes the Government's (licensee) rights in any copyright and the Government and its authorized representatives have unrestricted right to reproduce in whole or in part said article under any copyright secured by the publisher.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the USERDA.


los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87545

An Affirmative Action/Equal Opportunity Employer

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASS TRANSFER CHARGES INDUCED IN COAL BLOCKS DURING THERMAL PROCESSING

N. E. Vanderborgh, J. P. Bertino and D. N. Hopkins

Los Alamos Scientific Laboratory

Los Alamos, New Mexico, 87545

A B S T R A C T

In-situ processing of coals allows energy extraction from underground seams without usual societal and environmental costs of coal technology. Current concepts involve hot-gas underground processing to effect a partial oxidation of the coal, i.e., underground gasification. Modeling these processes requires comprehension of mass transfer mechanisms that drive the thermal transport processes within coal blocks.

Mass transfer is initially limited by the relatively low permeability of the natural occurring material. Pore geometries in coal suggest that mass transfer channels are near 50 nm. These pores are typically filled by absorbed moisture; moisture removal changes permeability by $10^{2.5}$. Once moisture is removed, other absorbed gases, CH_4 , CO_2 , etc., flush from the interior volume. These combined gases, during the drying steps, control preliminary heat transfer.

Modeling studies suggest that heat transfer mechanisms switch from conduction to convection as permeability is changed from 0.01 md to 5 md, those variations in mass transfer resistance formed during coal drying. Results are described that predict heat transfer rates in blocks of subbituminous coals during the initial drying stages of in-situ processing.

MASS TRANSFER CHANGES INDUCED IN COAL BLOCKS DURING THERMAL PROCESSING

by

N. E. Vanderborgh, J. P. Bertino, and
D. N. Hopkins

Los Alamos Scientific Laboratory
Los Alamos, NM 87545

INTRODUCTION The LASL concept for underground coal conversion is shown in Figure 1 (1,2). Coal, 250-1,000 feet underground, is first heated with hot gas injection using a downhole manifold inlet system. This heating produces moisture and hydrocarbons leaving in place a semi-char for subsequent gasification. The hot char, following the "precondition" stage, is gasified with a CO_2/O_2 mixture to produce a stream of low-Btu fuel gas (approximately 250-300 Btu/scf) for surface heat generation (electricity production). Before this fuel is utilized, the produced gas is cooled and then cleaned for sulfur and nitrogen removal. Sensible heat, stripped from the gasification products, drives the first stage of another coal section ("precondition"). Approximately 25% of the total heating value of the coal is produced as hydrocarbons removed during the initial pyrolysis stages.

This approach to underground coal conversion is based on the realization that, for efficient recovery, underground moisture control is essential. Long field experiences show that excessive moisture influx degrades underground gasification to, eventually, produce only small quantities of H_2 and CO , the desired fuel gases (3). We suggest that moisture removal, prior to processing, is sensible to increase both process efficiency and control. (Such removal would only be possible if additional water is excluded from the processing region once drying begins. Exclusion might well involve constructing underground barriers to prohibit water flows into the active zone. This possibility is not described here (4).)

Underground coal processing requires the control of a thermal wave moving through realistically large coal sections. Previous work on coal combustion has centered on finely ground material, the contemporary com-

mercial fuel. In those cases heat transfer by convective processes is unimportant. However, naturally occurring coal is a different situation. There is little question that such coal is fractured. Many seams are productive aquifers; water influx into underground workings has been troublesome through history. Coal also exhibits mineral inclusions, sites of ancient healed cracks. Evidence suggests that significant water flow occurs through large, millimeter-sized cracks. Water in these crack systems exerts fluid pressures within the entire coal mass including within the pore structures.

On a microscopic level coal is thought to consist of fine pores. Evidence, derived from dynamic gas adsorption behavior, suggests that pore geometries are of molecular size (5). Water is strongly adsorbed within such pore structures (6). Liquid water under the influence of pressure or temperature gradients drains to permit enhanced drainage rates for methane production or other gaseous species. These types of drainage are also important in underground coal processing.

Borehole permeability measurements within coal seams frequently show high liquid transfer rates. Flow occurs as the result of either natural or induced fractures since ample experimental evidence suggests that native, wet coal exhibits permeabilities near 0.05 md. Moisture removal and concurrent thermally induced crack formation greatly lowers mass transfer resistances. Since, during underground coal processing, the dominant heat transfer mechanism is convective transport (2), prediction of processing requires knowledge of these changes induced in transport properties during thermal treatment. This paper reports experimental data that describe such changes in coal and coal model materials that have contributed to our understanding about heat transfer rates through coals.

GAS TRANSFER THROUGH COAL It is often assumed that Darcy flow equations adequately represent mass transport in coal. However, flow measurements are complicated by sampling difficulties and by the fact that the process of mass transfer, by varying moisture distribution in coal, changes permeability. Moreover, natural coal is under considerable stress. Removing stress results in the formation of microfractures. Permeability measurements, consequently, need return coal sections to simulated in situ conditions.

Measurements underway study the influences of stress on mass transfer in coal. Apparatus for these measurements is shown in Figure 2. Coal samples, right cylinders cored from a freshly recovered sample, are confined in an elastic (neoprene) sleeve. This sleeve with sample is inserted into a tightly fitting brass cylinder. Geometries are so arranged that the elastic confining sleeve is slightly longer than the brass ring. The confining chamber is positioned between two platens in a hydraulic system. Transverse force is transferred through the neoprene elastic sleeve into radial confining stress on the coal sample. Sample gas, nitrogen, moisture or CO_2 , is transferred from one "ballast volume" to the other. Pressures are measured in a dual-differential mode so that gas volumes both into and out of the sample can be independently measured.

Results suggest that stress markedly changes gas permeability. Coal samples measured with no radial stress show permeability in the region of 40 to 100 md. However, examination of such samples showed the existence of microfractures. Tagged flow experiments show that the majority of flow transfer occurs through such cracked zones.

Data on permeability changes induced by stress loading are shown in Figure 3. Initial flow apparently is again through microfractures. Stress loading changes permeability from approximately 10 md to, at equivalent of 500 ft of overburden, 0.02 md. This factor of $10^{2.6}$ decrease in permeability is partially inelastic. Curve B, Figure 3, shows similar data following measurements given in Curve A. The coal, following removal of stress, does not return to the same configuration. This suggests that pressurization is sufficient to heal, at least temporarily, microfractures in coal. Since naturally occurring coals are stress loaded, we assume that most microfractures will be closed.

Data were also obtained following moisture removal. CO_2 gas flow was particularly effective in moisture removal. Moisture yields are followed by quantitative measurement of water recovered from a coal trap placed in the apparatus. Repeating results given in Figure 3 on partially dried samples (approximately 60% of the total moisture that would be removed by drying at 110°C was recovered at 25°C during CO_2 flow through), stress was again found to constrict flow. However, the effect was considerably less important. Dried coal, in the same pressure interval,

showed a permeability decrease of $10^{1.6}$.

These results show the marked changes in coal properties caused by stress-induced geometric variations. Pore geometries are of that size, that wet coal under simulated confining stress is impermeable. Moisture removal from the pore system leaves an open channel structure that is not readily collapsed by confining stress.

HEAT INJECTION INTO CERAMIC BLOCKS These results suggest that coal drying is the critical first step in underground processing. Not only is water transfer effective in moving heat within the coal mass, but, as described above, water plays a dominant role in determining mass transfer resistance. Laboratory experiments were designed to simulate drying under those conditions expected to occur during hot-gas injection (the "precondition" step in Figure 1).

The easily changed flow resistances in coal coupled with the variety of gaseous and liquid products resulting from heat injection, complicates modeling. Initial experiments were done using ceramic blocks, materials with well-described physical properties and reasonably simple chemistry. The first material selected was plaster of Paris ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). A right cylinder of this material was formed in a plexiglas mold holding a series of thermocouples. (In this way the thermocouples were imbedded in the block during fabrication.) A 0.125 inch rod, positioned in the center of the mold, was removed before the ceramic completely set. The resulting hole serves as a highly permeable path for heat introduction. The two ends of the cylinder were sealed so that the sole inlet into the block was the center hole and all gas exited from the center exhaust manifold. An inline heater was then attached to the system to inject heat at a rate determined by inlet temperature and gas flow rate.

Our task is to determine the temperatures at discrete positions within the ceramic as a function of time and then compare those experimental results with a combined heat and mass transfer code (2) to learn if such hot gas injection results in predicted thermal excursions. Results presented previously suggest that such hot gas heating in semi-porous media can be adequately predicted if boundary conditions are well described (2). Such experiments assume a constant and uniform permeability throughout the block.

Clearly, this condition would not occur in drying coal blocks. In that case moisture transfer will alter permeability to direct flows into particular regions and exclude flow from others. Although it is possible to introduce moisture into the interior of ceramics, the removal of such moisture with hot gases involves a complicated two-phase, two-component flow. Simpler systems involve steam injection into a block that is slightly lower than equilibrium steam temperatures so that condensation occurs. This is the reverse of the in situ condition. However, once the computer code adequately predicts the "wetting" experiments, then it is straightforward to reverse the code to predict "drying." Although we are encouraged with these variable permeability modeling codes, computer results are not presented here (7).

Results of steam injection into the ceramic block are shown in Figure 4. Liquid water is injected into the inline heater (maintained at 120°C) at a rate of 0.40 ml/min. Steam generated within the heater flows into the center channel and out into the block. Concurrently, temperatures are measured at fixed locations. Initially, the entire block is maintained at 80°C within the thermostat. At the onset of flow thermocouple one, located in the entrance channel, measures a sharp temperature rise. As the steam flow continues, temperatures at the exhaust side of the block increase. This result is completely opposite from that found with CO₂ injection in dry materials (2).

After approximately 200 ml of water is injected into the ceramic, the water stream is again switched to hot CO₂. Now gas transfer occurred through the inline heater (175°C) at 300 ml/min. Data are shown in Figure 5. Here again the interior block temperatures are shown as well as the inlet temperature (Number 1) and the exhaust stream (Number 8). The thermal wave heats the inlet side of the block, moving along the block as time progresses. During the first 24 hours of flow, gas flow results in cooler temperatures than those initially measured in the interior. Finally, the last curve in Figure 5 shows that, once moisture is removed, heating of the block is much like that described with CO₂ injection in dry ceramics (2).

These data are predicted if we assume that mass transfer in unique regions of this ceramic cylinder is controlled by local moisture content.

Dry and permeable regions allow efficient heat transfer, as shown previously, by convectively moving hot gas from the central channel into the interior of the block. In the dry situation the inlet side of the block reaches highest temperatures and the exhaust side, due to various heat losses, remains at ambient (furnace) temperature. Steam injection changes the permeability in those zones near the inlet side of the central channel. This zone immediately near the channel accepts heat by steam condensation. However, due to this moisture deposition, these zones are sealed to additional flow. Heat transfer in the wet zones is retarded (ceramics and coals have slow heat conduction rates) and heat appears at the exhaust end of the block. Transfer occurs not only by steam down the central bore, but also by hot gases excluded from the volume occupied during moisture condensation.

Reinjection of hot gas into the wet block results in evaporative cooling. Inlet CO_2 evaporates moisture in unique zones. This process cools that zone below the initial inlet temperature. Data show that cooling occurs rapidly after gas flow is started and continues for extended time periods maintaining lower temperature than either the ambient thermostat or inlet gas temperature.

HEAT INJECTION INTO COAL BLOCKS Ongoing experiments repeat these studies with instrumented coal blocks. Conveniently sized sections of subbituminous coal are bounded with mass transfer barriers so all flow into the block must enter in one position. Likewise, flow exhausts from the block in one channel. Thin (28 gauge) thermocouples are then inserted and the holes are back filled with cements. Likewise, thermocouples are inserted into the inlet and exhaust sections of the central channel (previously drilled with a 0.125 in bit). The thermocouples are connected to recording equipment, inlet heat connected, and heat injection commences.

Coal, during drying with hot CO_2 , yields thermal results most like those obtained with the wet ceramic block. Data appear in Figure 7. As previously, temperatures at specific sites within the block are shown after two different heat injection rates. Measurement positions are determined by x-ray examination of a block prior to the heating experiment. The lower curve was determined first. The inlet side increased in temperature while the center section decreased in temperature. (The entire block was initially at 100°C.) Finally, after extended moisture removal, the block

yields data similar in thermal behavior to that noted with dry ceramic materials.

We interpret these data as illustrating enhanced flow through coal sections resulting from thermal treatment and moisture removal. As the thermalized zone progresses outward, the heated region increases both outward from the central channel and along the axis of the coal block. Evaporative cooling is also clearly observed. Gas injection in the native material causes evaporation and, initially, a decrease in temperature. (The sign of temperature changes depends upon the relative rates of heat introduction and removal.)

CONCLUSIONS These data coupled with heat and mass transfer modeling (not presented here) begin to support a conclusive insight into coal drying. Permeability in coals is markedly a function of stress and fluid saturation. In virgin materials the greatest majority of flow is conducted along regions of naturally high permeability, cracks, for example. Although this sort of flow is indeed significant for water production, flow through such channels bypasses major regions of the underground seam. Moisture removal changes this situation.

Permeable coals readily transfer heat into the coal mass by convective transport processes. Such open systems can be rapidly sealed again by the introduction of additional moisture. During underground coal processing moisture removal is that key factor that permits processing zones to broaden into wide regions that permit efficient resource recovery, much as found during early Hanna experiments. Thus, even though moisture content can adversely influence processing schemes, moisture control, technically feasible, would permit the control of underground flow to recover with good efficiency deeply lying coal seams. That goal is the thrust of the LASL effort in underground coal conversion.

REFERENCES

1. N. E. Vanderborgh, E. M. Wewerka, W. J. Trela, J. M. Williams and G. R. B. Elliott, "Combined CO₂-O₂ Underground Pyrolysis-Gasification for Southwestern Coals", preprint, Fuels Division, American Chemical Society, presented at the Joint ACS-Canadian Institute of Chemistry Meeting, Montreal Canada, June 1, 1977. LA-UR-77-696
2. N. E. Vanderborgh, E. M. Wewerka, J. M. Williams, J. P. Bertino and G. E. Cort, "Heat and Mass Transfer Through Southwestern Subbituminous Coals", presented at the Third Underground Coal

Symposium, Fallen Leaf Lodge, Lake Tahoe, CA, June 1977, LA-UR-77-1341.

3. D. W. Gregg, "Critical Parameters of *In Situ* Coal Gasification (California University, Livermore (USA), 5 Feb. 1975, UCRL-76-496; A. A. Agroskin, T. W. Sukhotinskaya and N. A. Fedorov, "Moisture Balance in Underground Gasification; Podzemnaya Gazifikatsiya Uglei 1, 25 (1958); G. O. Nusinov, N. Z. Brushtein and M. A. Kulakova, "Gasification of Brown Coal Underground by Preheating the Coal Seams and Removing the Gas in Two Stages", *ibid*, 3, 16, (1959).
4. S. A. Colgate, Los Alamos Scientific Laboratory, personal communication. (January 1978).
5. J. Thomas, Jr., H. H. Damberger, "Internal Surface Area, Moisture Content and Porosity of Illinois Coals: Variations with Coal Rank, Illinois State Geological Survey, Circular 493, 1976.
6. P. Cinche, H. Marsh and S. Pregermain, "Absorption of Carbon Dioxide, Methanol and Water Vapor on Cokes, Determination of Micropore Volume," *s Fuel*, 46, 341, 1967.
7. N. E. Vanderborgh, G. R. B. Elliott, G. E. Cort, "Concurrent Heat and Mass Transfer During Drying Of Blocks of Subbituminous Coals", Fourth Annual Underground Coal Conversion Symposium, July 17-20, 1978, in press.

FIGURES

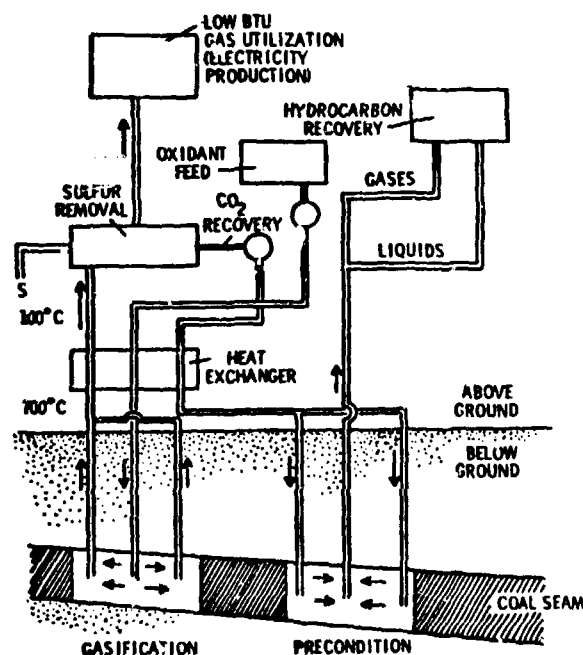


Figure 1: Schematic of Proposed Underground Pyrolysis-Gasification Process. Sensible heat from char gasification is introduced virgin seam causing drying and pyrolysis. Pyrolysis removes hydrogen enriched products leaving char behind for gasification.

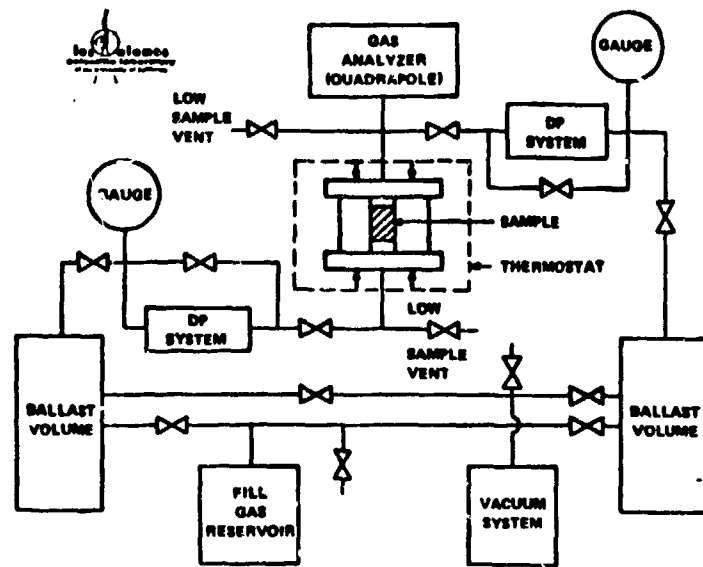


Figure 2: Schematic of Laboratory Apparatus to Measure Influence of Confining Stress on Mass Transfer Rates in Coal Samples. Dual-differential pressure monitoring permits separate monitoring of gas flux in and out of sample cylinder.

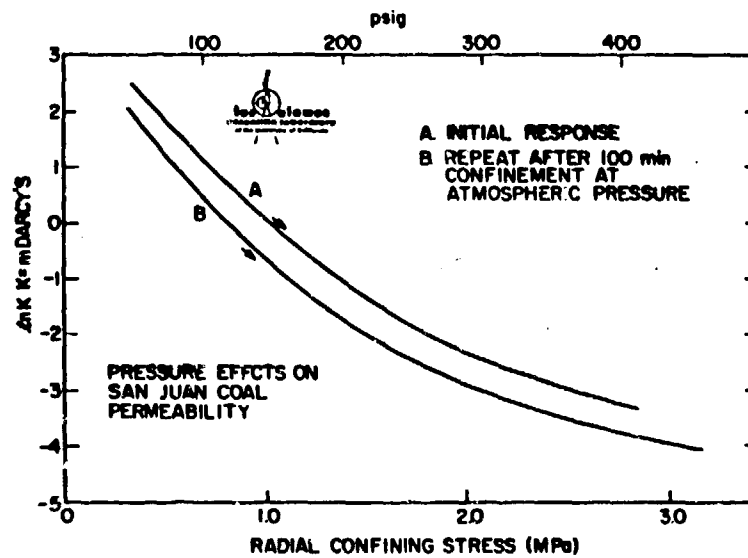


Figure 3: Effects of Confining Stress on Coal Permeability. Sample removed from Sage Pit, Fruitland Seam (Western Coal Company, Farmington, NM). Sample orientated so that flow is parallel to bedding. Curve A shows initial response; Curve B shows response of subsequent runs. 25 C° data.

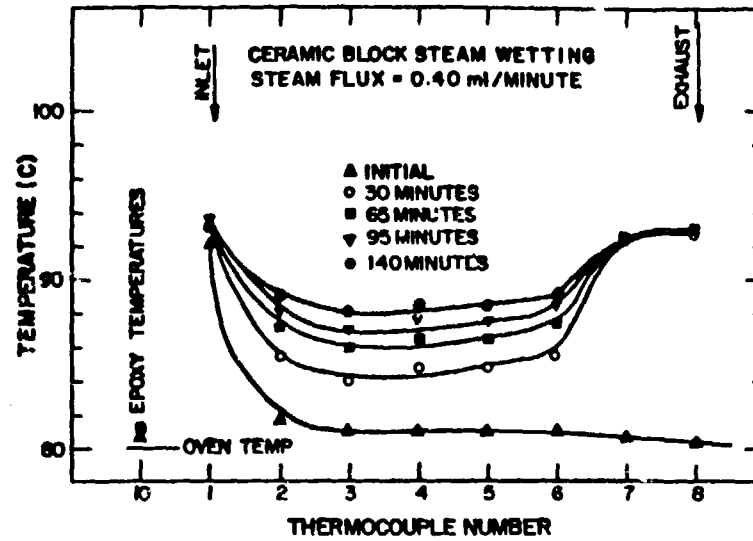


Figure 4: Thermal Response of Ceramic Block During Steam Wetting (Inverse Drying) Experiments. Thermocouple number approximately describes location of temperature. Block dimensions, approximate, 10 in (25.4 cm) long, 8 in (20 cm) diameter, 0.125 in (0.32 cm) diameter central channel (open hole). Thermocouple 10 measures temperature in the inlet stream and 8 measures temperature in the exhaust stream.

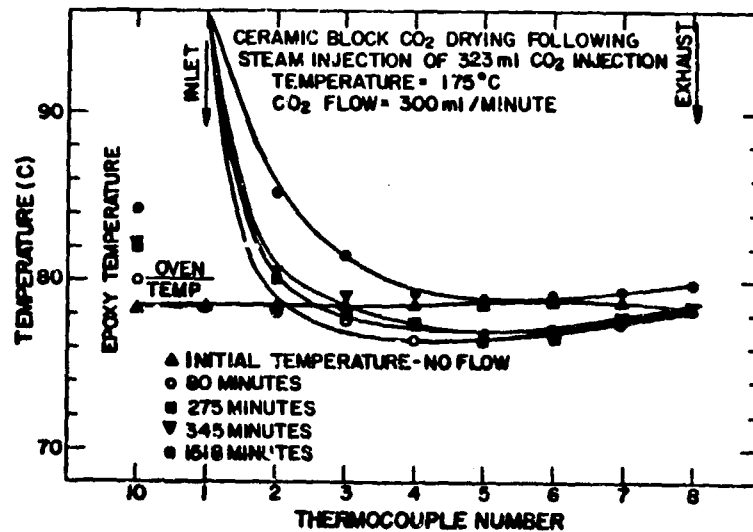


Figure 5: Thermal Response of Ceramic Block During Hot Gas Drying Experiments. Hot CO₂ injection following wetting experiment shown in Figure 4.

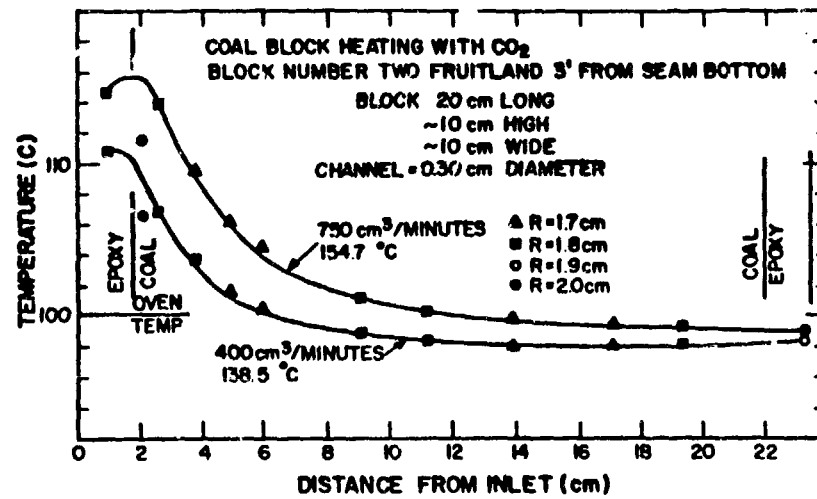


Figure 6: Thermal Response of Coal Block. Coal removed from Fruitland Seam, sectioned and cast into epoxy to provide complete mass transfer boundary. Blocked drilled (0.125 in hole) through center and gas injection occurs (only) through center channel. Entire block is first heated (no flow) to 100°C and then flow initiated. Two separate data shown.